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REVIEW ARTICLE

Biofouling in reverse osmosis: phenomena, monitoring, controlling and remediation

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Abstract This paper is a comprehensive review of biofouling in reverse osmosis modules where we have discussed the mechanism of biofouling. Water crisis is an issue of pandemic concern because of the steady rise in demand of drinking water. Overcoming biofouling is vital since we need to optimize expenses and quality of potable water production. Various kinds of microorganisms responsible for biofouling have been identified to develop better understanding of their attacking behavior enabling us to encounter the problem. Both primitive and advanced detection techniques have been studied for the monitoring of biofilm development on reverse osmosis membranes. Biofouling has a negative impact on membrane life as well as permeate flux and quality. Thus, a mathematical model has been presented for the calculation of normalized permeate flux for evaluating the extent of biofouling. It is concluded that biofouling can be controlled by the application of several physical and chemical remediation techniques.

Keywords Biofouling · Reverse osmosis · Mechanism · Control · Consequences · Disinfection · Surface modification

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Introduction

Worldwide demand for drinking water is increasing rapidly. The world's population tripled in the twentieth century and is expected to increase by another 40–50% by 2050. Hence, improving the performance of water purification technology is necessary to compensate for our fresh water demands (Kang and Cao 2012). Reverse osmosis (RO) has become a critical technology in purification of non-traditional water sources such as brackish, sea, and wastewater and it is the most efficient technique for seawater desalination purposes (Matin et al. 2011). Around 20% of the world's population lacks safe drinking water. It is expected that by 2025, 1.8 billion people will find difficulties in getting clean water or will live in areas where water is scarce. Consequently, ensuring high performance of RO plants is important and this is possible by adjusting parameters like feed pressure, permeability, system temperature, flow rates, feed salinity, and controlling biofouling issues. Selecting the accurate operating conditions will allow us to determine the necessary membrane area and therefore reaching the optimum values for permeate water flux and salt rejection. For instance, applying a high pressure (ΔP) that is larger than the osmotic pressure ($\Delta \pi$) across the membrane, results in an increase in water flux and salt rejection (Qureshi et al. 2013). The most commercially available RO membrane is the asymmetric cellulose type (cellulose acetate, triacetate, cellulose diacetate or their blend) and thin-film composite (TFC) type. TFC aromatic polyamide membrane exhibits superior water flux and salt rejection (Kang and Cao 2012).

Fouling occurs when dissolved and particulate matter in feed water deposits on the membrane surface leading to an increase in the overall membrane resistance (El Aleem et al. 1998). In other words, fouling happens when solutes in the flow are adsorbed reversibly or irreversibly onto the membrane surface or within the pores of the membrane. The irreversible adsorption is the main issue and it produces a long-term flux decline (Matin et al. 2011). There are four categories for fouling sources (as seen in Table 1): scale (inorganic), particulate, biological and organic compounds. Biofouling depends on the amount of biological, organic matter and colloidal particles in the feed water. Eliminating these particles (through pretreatment) in feed water is the main objective to avoid major biofouling problems in the final RO modules of the plant that are the most affected elements. Another effective way to increase the recovery rate is to have a partial membrane replacement (Qureshi et al. 2013).

Saudi Arabia produces around one-third of the world's capacity of desalinated water. Current desalination technologies in the Kingdom of Saudi Arabia include multistage flash method (MSF) and the RO process. RO process is preferable since it is simple, inexpensive and easy to maintain. However, recent critical problems related to RO membrane processes are fouling, biofouling, and biocorrosion (El Aleem et al. 1998).

Gulf water is rich in microorganisms, organics and has a high level of total dissolved solids (TDS) (>40,000 ppm). Thus, the main reason for flux decline in RO plants in the Middle East is biofouling. Biofouling reduces actual membrane performance through microbial generation in a biofilm which is formed on the membrane surface. Wastewater recirculation in industrial treatment plants results in having a higher concentration of TDS that promotes bacterial growth and biofilm development. Further, the use of activated carbon system (GAC or PAC) before the RO modules increases biological fouling. Hence, proper pretreatment, disinfection, and micron cartridge filters are important to control bacterial growth during RO treatment process (El Aleem et al. 1998). Reducing the concentration of microorganisms and nutrients in the feed to the RO membrane, adjusting the properties of the RO feed water and removing the developed biofilm on RO membrane can be regarded as some other approaches that could be applied to solve the problem of biofouling in RO modules.

Biofouling in a seawater reverse osmosis (SWRO) plant is controlled by the surrounding environment as well as pretreatment of feed water. The population of bacteria in seawater is dependent on various environmental factors such as light, temperature, tides, currents, turbidity and nutrients. SWRO module is more vulnerable to biofouling in hot climatic conditions. For example, degradation of humic acid is much easier and greater at a temperature of 35 than 18 °C. Degraded small molecules are a source of nutrition for bacterial growth. Since RO feedwater and brine reject temperatures are always higher than that of seawater feed, a higher biofouling potential is expected at the increased operation temperature. In addition, water samples near shore surface at Al-Birk plant in Saudi Arabia showed less nutrient content than water samples from the intake. It is important to choose an intake site that is less in nutrients and silt to avoid biofouling since the water source may have a negative impact on the operation parameters. Studies showed that the shortest bacterial growth generation time is ~ 2.5 h meaning that biofouling is a biofilm problem. RO membranes have an enormous surface area that increases the chances of a single bacterium to reach a membrane surface and later colonize to form a biofilm (Saeed et al. 2000).

Biofouling causes severe losses in performance of RO membranes and requires costly cleaning procedures to remove biofilms. Impact of biofilms on plant performance is linked to the structure and composition of the biofilm. Microorganisms including bacteria are the main reason for biofouling and since bacteria is very adaptable, it is capable of colonizing almost any surface at extreme conditions such as temperatures from -12 to 110 °C and pH values between 0.5 and 13 (Qureshi et al. 2013). Table 2 shows the most common microorganisms that can attack RO membranes.

Table 1 Types of fouling in RO membrane systems (Qureshi et al. 2013; Kang and Cao 2012)

Fouling type	Mechanism	Causing substances
Inorganic	Deposition of inorganic materials	Metal hydroxides, carbonates, sulfates, phosphates
Organic	Deposition of organic substances	Oil, proteins, humic acids, polysaccharides, lipids
Particulate and colloidal	Deposition of debris and other substances	Clay, silt, silica
Biofouling	Adhesion and accumulation of microbes, forming biofilms	Bacteria, fungi, yeast

Table 2 Common microorganisms identified in biofilms (Qureshi et al. 2013; Baker and Dudley 1998)

Bacteria	*Mycobacterium, Flavobacterium, Pseudomonas, Corynebacterium, Bacillus, Arthrobacte, Acinetobacter, Cytophaga, Moraxella, Micrococcus, Serratia, Lactobacillus, Aeromonas*
Fungi	*Penicillium, Trichoderma, Mucor, Fusarium, Aspergillus*
Yeasts	Occasionally identified in significant numbers

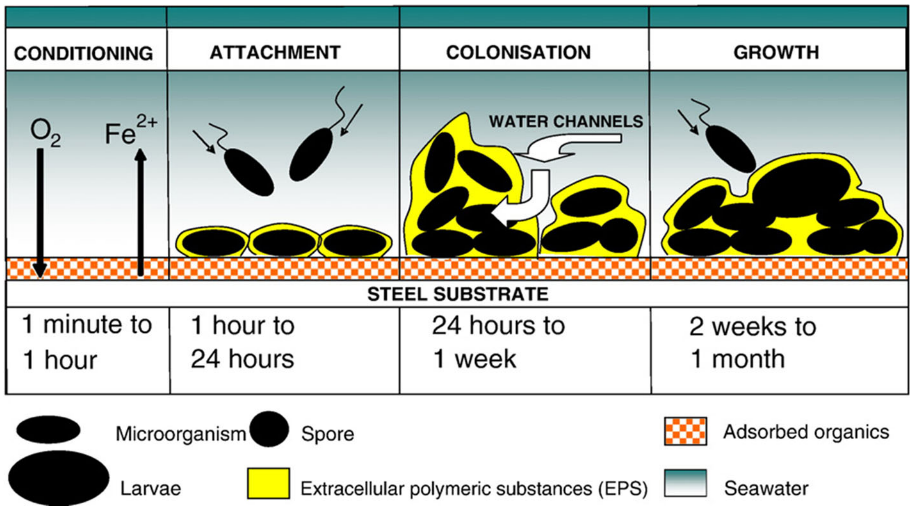


Fig. 1 Mechanism of biofilm development

Biofilm development

Mechanism

Biofouling process or biofilm formation is a multistage process that is complex, slow, reversible or irreversible process where microbial growth can take couple of weeks or months. However, the initial step (adsorption) is relatively fast and can occur in about 2 h only. Mechanism of biofilm development is illustrated in Fig. 1. Biofouling process goes sequentially through the following steps (Matin et al. 2011; El Aleem et al. 1998).

1. Adsorption of organics onto the wetted membrane surface (conditioning): Biofouling occurs through a cascade of events including the transport, deposition and adhesion of cells followed by exopolymer production, cell growth and proliferation. Conditioning enhances attachment of cells to the surface.
2. Transport and attachment of the microbial cells to the conditioned surface: This step depends on different physical and chemical factors, but attachment generally is more favorable with hydrophobic, non-polar surfaces.
3. Growth (metabolism) of the attached microorganisms and biofilm development: Biofilm formation stage takes place by auto-aggregation of the attached cells and formation of microcolonies. Extracellular polymeric substances (EPS) are continuously produced and acts as a reactive transport barrier to chemical biocides and promotes nutrient concentration/storage.
4. Detachment and limitation of biofilm growth by fluid shear forces: Cell detachment is an active form of dispersion of cells from the biofilm matrix and detached biofilm cells reinitiate biofilm formation on new sites. Understanding this step is important since it is related to the control of growth.

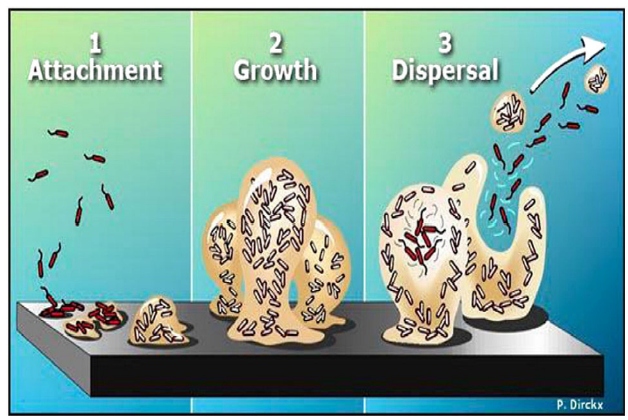


Fig. 2 Sequence of events leading to the formation of a Biofilm (Cunningham et al. 2011)

The primary induction phase is followed by the logarithmic growth phase which contributes more to microbial growth as compared to microbial adhesion; then plateau phase which is mainly controlled by the presence of nutrients. When plateau phase is attained, the membrane is masked by the biofilm (Matin et al. 2011). More details about each phase are summarized in Fig. 2 and below (Flemming 1997).

Induction phase refers to the primary colonization of the membrane by microorganisms. The primary colonization is followed by a primary plateau. The induction phase also refers to the time between two cleaning measures. Colonization takes place due to microbial adhesion which is proportional to the cell density in the water phase and occurs owing to weak physicochemical interactions (Flemming 1997).

Logarithmic phase involves cell growth which contributes more to biofilm accumulation than adhesion of planktonic cells (Flemming 1997; Schaule 1992).

Plateau phase is governed by nutrient concentration and the resultant growth rate, mechanical stability of the biofilm, and effective shear forces. It is independent of the concentration of cells in the feed water. In this phase, we have another plateau which represents the balance between biofilm growth and cell detachment. The concentration of assimilable organic carbon is the key parameter controlling the level of the plateau which is significant for process stability, energy consumption, and economics (Flemming 1997).

Threshold of interference in Fig. 3 is the extent of biofilm development above which the biofilm interferes with the performance of a membrane system. Treatment techniques focus on getting the microbial concentration levels beneath the defined threshold of interference (Flemming 1997).

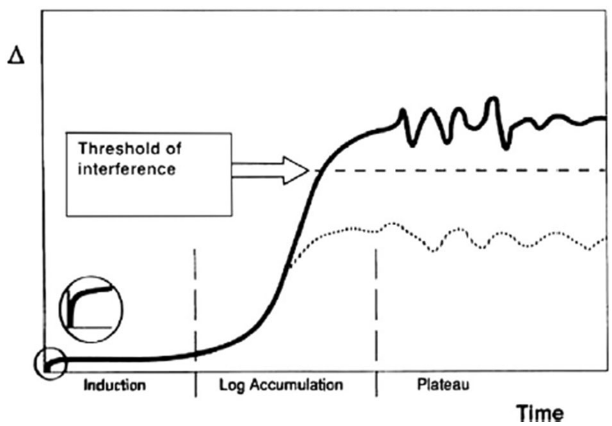


Fig. 3 Development of biofilm and accumulation of microbial matter with respect to time (Flemming 1997)

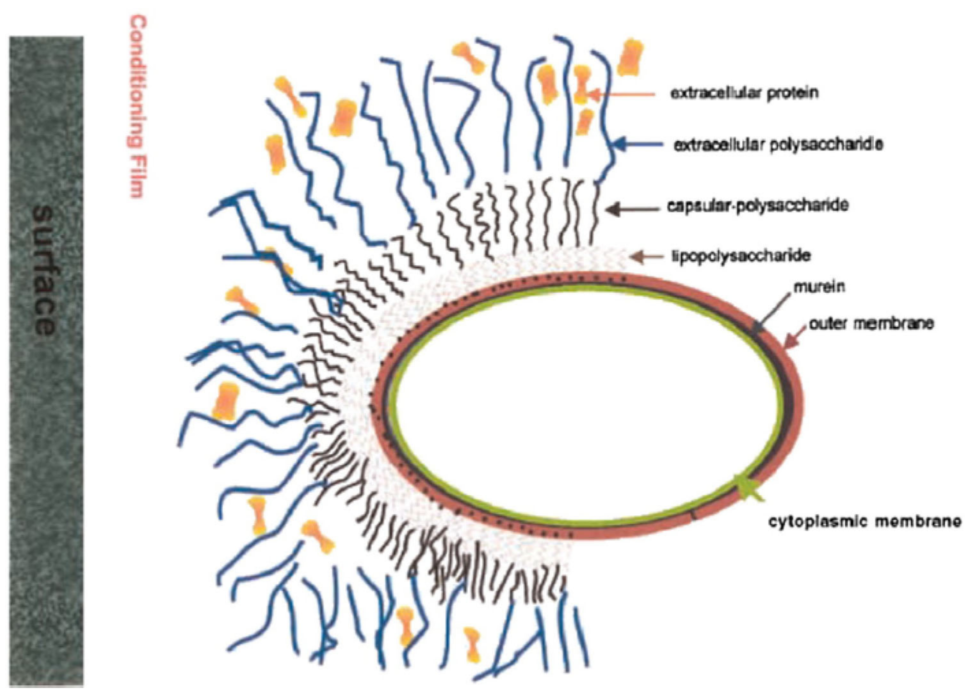
Biofouling occurs due to the deposition and growth of biofilms. However, biofilm generation starts when the attached microorganisms excrete EPS. Biofilms are composed primarily of microbial cells and EPS as shown in Fig. 4. EPS constitutes 50–90% of the total organic carbon (TOC) of biofilms and is considered as the primary matrix material of the biofilm. EPS consists primarily of polysaccharides, proteins, glycoproteins, lipoproteins, and other macromolecules of microbial origin. The EPS matrix offers important advantages for bacteria like maintaining stable arrangements of the cell and enhancing the degradation of complex substances (Matin et al. 2011).

Factors influencing microbial adhesion

Transport conditions play an important role in microbial adhesion as they affect the accumulation of microorganisms on the surface of the membrane. These transport conditions also influence generation of shear forces. High shear forces are desirable as they inhibit microorganism adhesion and hence microbial growth at the membrane surface (Al-Juboori and Yusaf 2012).

pH of solution affects the electrostatic double layer interaction between the membrane and microorganisms due to change in surface charge. Change in pH of the solution has a slight effect on the surface charge of the membrane but has a substantially higher effect on colloids' charge (Brant and Childress 2002).

Fig. 4 EPS components of a bacterium encountering a non-biological surface in water (Tamachkiarow and Flemming 2003)



Ionic strength of solution also affects the electrostatic double layer interaction between the membrane and the microorganisms. Most microorganisms are negatively charged; so in order to avoid microbial adhesion and subsequent growth on the membrane surface we desire that the membrane should also be negative thereby inhibiting adhesion due to repulsive forces (Al-Juboori and Yusaf 2012; Lee and Elimelech 2006; Hong and Elimelech 1997).

The characteristics of interacting surfaces that play a significant role in biofilm formation are hydrophobicity, hydrophilicity, and surface roughness. Hydrophobicity and hydrophilicity are analogous properties that determine the membrane's tendency to foul. As the name suggests, hydrophobic membranes preferentially interact with microbial matter which causes biofouling; while hydrophilic membranes interact with water. Another crucial factor is surface roughness of the membrane. Rough surfaces have larger number of sites convenient for microbial adhesion in the form of peaks and troughs. Rough surfaces also have larger surface areas than smoother surfaces thereby increasing the number of sites for adhesion. Moreover, the roughness of the membrane surface can decrease the Lifshitz–van der Waals and electrostatic double layer interactions of the membrane (Brant and Childress 2002; Yu et al. 2010).

Nutrients in the bulk solution serve as food for microorganisms; hence, concentration of nutrients should be low to avoid biofouling. While the presence of nutrients is not directly detrimental to the membrane, it acts as a source of nutrition for microorganisms aiding their metabolic activities and growth. It has been found that increasing the concentration of carbon in bulk solution, shortens the initial growth period of the biofilm resulting in lesser microbial mass (Al-Juboori and Yusaf 2012).

Higher concentration of microorganisms in the bulk solution leads to higher adhesion and microbial growth on the membrane surface as well as higher generation of EPS which fouls the membrane and reduces membrane flux (Al-Juboori and Yusaf 2012). Factors affecting bacterial multiplication rate are feed water quality, temperature, pH, dissolved oxygen content, the presence of organic and inorganic nutrients, pollution, depth and location of the intake (Saeed et al. 2000; El Aleem et al. 1998).

Moreover, biofilm development is also influenced by the carbon: nitrogen: phosphorus ratio, and redox potential. Physical structure of biofilm can be compact and gel like or slimy and adhesive with large amounts of polysaccharide. Generally, biofilm contains between 10^6 and 10^8 colony forming units (CFU) of bacteria per cm^2 of membrane area. There is a strong relation between biofilm composition and various environmental factors such as temperature and humidity. In Table 3, we have a typical biofilm composition from previous laboratory studies for brackish and seawater treatment plants:

Table 3 Typical composition of biofilm (Baker and Dudley 1998)

Parameter	Composition
Moisture content of dried deposit	>90%
Total organic matter (TOM)	>50%
Humic substances as % of total organic matter	≤40%
Microbiological counts	$>10^6$ cfu/cm^2

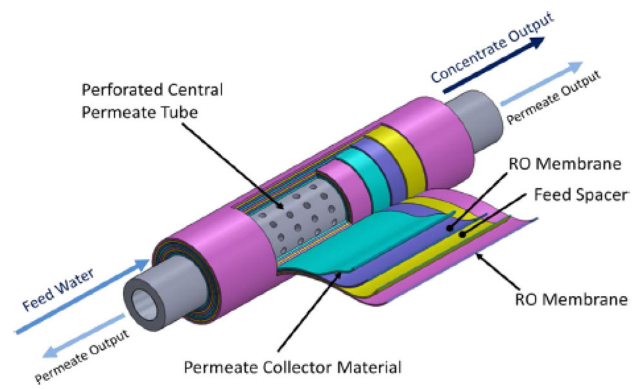


Fig. 5 Spiral-wound RO module (Qureshi et al. 2013)

Reverse osmosis module

Biofouling in RO module elements include the formation of biofilms in permeate surfaces of cross-flow membranes, woven polyester support fabrics, permeate collection material, and feed channel spacer materials. The crucial biofouling type in RO module is the formation of biofilm in the feed channel spacer material. This should be avoided to restrict the impact of biomass accumulation on the feed channel pressure gradient increase. Fig. 5 represents a spiral-wound RO module.

The spacer minimizes the problem of concentration polarization since it consists of a network of plastic fibers that separates the spiral wound membrane sheets from each other to create turbulence and inhibit further biofouling. Channeling problems happen in hollow fiber bundles when we have individual fibers that are bounded together which causes rapid salt concentration leading to the precipitation of salts such as calcium carbonate and calcium sulphate (Matin et al. 2011). Table 4 summarizes bacteria counts in biofouled systems that produce potable water (Baker and Dudley 1998).

Table 4 Typical microbial activity in biofouled spiral wound elements (Baker and Dudley 1998)

	Range of viable bacteria counts (cfu/cm^2)	Range of fungal counts (cfu/cm^2)
Fouled membrane	1×10^2–1×10^8	0–1×10^3
Plastic spacer material[a]	4×10^2–5×10^6	0–1×10^3
Permeate carrier	$<10^2$–1×10^6	None

[a] Viable bacteria computed per cm^2 of the spacer mesh

Modeling and monitoring

Modeling of flux decline

In RO systems, the most important parameters in terms of design and performance are the feed pressure and feed concentration, respectively. A solution-diffusion model for steady-state processes showed a good agreement between the experimental or measured results and simulated results (Qureshi et al. 2013).

Fouling analysis model with two constants is proposed for predicting the normalized decrease in permeate flux due to fouling. Membrane fouls over time and fouling curve exhibits an asymptotic behavior. Fouling of RO membranes can be modeled using a normalized permeate flux decline η_J that follows the following relation and varies with time (Khan and Zubair 2004; Qureshi and Zubair 2005).

$$\eta_J = \eta_J^*[1 - \exp(-t/\tau_c)] \quad (1)$$

where η_J^* is the asymptotic value of the normalized permeate flux decline (η_J) and τ_c is the time constant expressing the time when the normalized permeate flux (η_J^*) reaches 63.2% of its asymptotic value. η_J^* and τ_c are two constants to be determined beforehand. This model is used to predict the decrease in permeate flux as the membrane fouls over time. Literature shows that both constants depend on the feed concentration, cross-flow velocity, pH and transmembrane pressure drop (Qureshi et al. 2013; Khan and Zubair 2004; Qureshi and Zubair 2005).

$$\tau_c = f(C_o, u, \Delta\mathrm{P}, \mathrm{pH}, T) \quad (2)$$

$$\eta_J^* = f(C_o, u, \Delta\mathrm{P}, \mathrm{pH}, T) \quad (3)$$

Koltuniewicz and Noworyta (Koltuniewicz and Noworyta 1994) suggested two equations for the calculation of both constants as follows:

$$\frac{1}{\tau_c} = 0.298 \times 10^{-4} C_o^{0.567} \Delta\mathrm{P} \quad (4)$$

$$\eta_J^* = \frac{3.875 \times 10^{-6}}{C_o^{1.21}} \quad (5)$$

However, authors reported a maximum relative error for Eqs. (4) and (5) which is about –13.1 and –20.1%, respectively. Since we have large error values, they can not be neglected; further investigations and experimental works are needed to determine accurate constant values for specific RO applications. Practically, integration of the model into an RO cleaning strategy helps in identifying the affected membrane points and whether a backwash with or without cleaning chemicals is needed or not (Qureshi et al. 2013). Fig. 6 demonstrates the normalized decrease in flux of permeate with respect to time for different feed pH values.

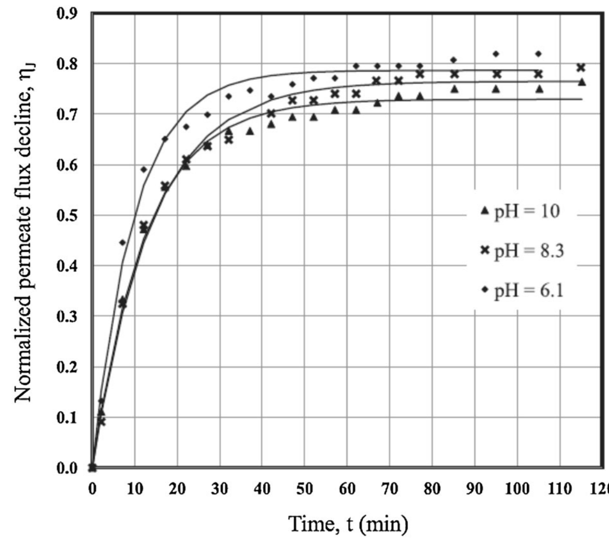


Fig. 6 Curve fit of normalized permeate flux decline versus time (Qureshi et al. 2013)

Monitoring and detection

The first step towards addressing biofouling through treatment is to detect formation of biofilms and monitor cell accumulation. Techniques by which this is done can range from primitive inspection through sight or smell, sampling and lab testing to more advanced techniques like bioluminescence, epifluorescence microscopy, etc. Here we will discuss the various techniques employed for detection and monitoring of biofouling (Al-Juboori and Yusaf 2012).

1. Physical inspection: RO systems such as the spiral wound membrane module may show signs of biofouling in smell and color which can be physically inspected. Routine visual inspection of various plant components such as pretreatment piping, cartridge and media filters should be done to detect accumulation of biological matter. All of these inspections must be performed in wet conditions since microorganisms thrive in it (Al-Ahmad et al. 2000).
2. System performance analysis: EPS secreted by microorganisms cause a decline in membrane flux.

The measurement of this change in flux and pressure drop across the membrane is a very good way of monitoring biofouling. Performance of the module is gauged by measuring the flow rate and purity of permeate, salt rejection efficiency, and silt density index (SDI) of feed water entering the module (Al-Ahmad et al. 2000).

3. Water sampling: Routine collection of feed, permeate and retentate streams should be done right from the onset of operation of RO plant. The sampling points should be chosen as to adequately cover the entire system. This monitoring technique primarily serves as a preventive measure. The main objective of this sampling and analysis technique is to locate or isolate the source of any bioactivity before it starts to spread and affect other parts of the RO system. Presence and accumulation of different species of microorganisms is measured along with SDI, pH, COD, TOC, and dissolved oxygen content. SDI is a measure of fouling potential; clean brackish water will have SDI <5, whereas, seawater will have SDI values ranging 6–20 (Al-Ahmad et al. 2000; Abd 1998).
4. Culturing techniques: These are employed to detect the kind of microbial activity as well as the concentration of those species affecting the RO system. Methods usually used for this biological analysis are either for measuring the total accumulation of biological matter or for the detection of specific species of microorganisms through analysis of microbial activity on cultured samples. Cultures are retained for 24–72 h at 25–30 °C (Al-Ahmad et al. 2000).

Table 5 summarizes most of the microscopic and spectroscopic techniques used for the inspection of biofilms in reverse osmosis modules. While each technique has its own advantages and disadvantages, Hoffman modulation contrast microscopy (HMCM) can be considered as the single most beneficial microscopic technique for monitoring of biofilm formation. HMCM (Fig. 7) has no significant drawbacks and has plentiful advantages. Being non-invasive, HMCM technique does not interrupt normal RO plant operation and trumps most other techniques by offering high resolution imaging without the need of preparation of any specific kinds of samples (Al-Juboori and Yusaf 2012).

Similarly, the authors believe that Fourier transform-infrared (FT-IR) spectroscopy is arguably the best spectroscopic technique to study the physiological behavior of microorganisms. FT-IR spectroscopy (Fig. 8) is the most commonly used spectroscopic technique as it not only detects microbial presence but can also distinguish between live and dead cells, thereby, aiding the subsequent controlling and treatment techniques. Moreover, biofilms can be in different phases and physical forms such as solid, colloidal or slimy films. Applicability of FT-IR spectroscopy irrespective of the physical nature of biofilm makes it the best spectroscopic technique for monitoring of biofouling (Brant and Childress 2002).

While FT-IR spectroscopy has drawbacks, the authors believe that these do not have any consequences on the legitimacy of this technique for monitoring of biofouling in RO systems. Since routine sampling is conducted to detect early onset of biofilm formation, the microbial growth and EPS secretion is highly unlikely to be significant enough to form a biofilm which is thicker than the order of 1 μm (Flemming 1997).

Furthermore, even though FT-IR spectroscopy requires a library of spectra for each microorganism for its identification after detection, owing to the culturing techniques discussed earlier, we already know the different kinds of microorganisms that are present in the feed. Hence, we need information on spectra of only those microorganisms which are present in the feed to the RO membrane and can potentially cause biofouling.

This analysis of drawbacks presents the conclusion that FT-IR spectroscopy is the best spectroscopic technique for monitoring of biofouling in RO systems as routine sampling of feed and culturing techniques can eliminate the disadvantages associated with this technique.

Consequences of biofouling

Biofouling has diverse consequences on the entire RO module, particularly the membrane system. It affects both the process as well as physical components of RO module. These effects are elucidated below (Baker and Dudley 1998; El Aleem et al. 1998; Flemming 1997).

1. Membrane flux decline: This is because of the formation of a film of low permeability on the membrane surface.
2. Membrane biodegradation: Microorganisms produce acidic byproducts that damage RO membrane.
3. Increased salt passage: Accumulated ions of dissolved salts on the membrane surface enhances concentration polarization and inhibits convectional transport.
4. Increase in the differential pressure and feed pressure: This is due to biofilm resistance.
5. Increased energy requirements: High-pressure requirements are due to higher feed pressure, frictional energy losses and drag resistance to tangential flow over the membrane.
6. Frequent chemical cleaning: Imposes a large economic burden on RO membrane plant operation, up to 50% of the total costs, and shortens membrane life.

Table 5 Microscopic and spectroscopic techniques for the detection of biofouling in RO membranes (Al-Juboori and Yusaf 2012; Khan et al. 2010; Wolf et al. 2002; Griffiths and De Haseth 2007; Chambers et al. 2006)

Technique	Advantages	Disadvantages
Microscopic techniques		
Epifluorescence microscopy	Rapid analysis, provides information on the structure–function relationships in biofilm	Unable to measure the depth of the biofilm, low resolution and the requirement of removing the biofilm (invasive technique)
Electron microscopy	Produces images with high resolution, and provide cross-sectional details of the biofilm, which allows visualizing the spatial distribution of microorganisms in the biofilm matrix	Unable to study biofilm structure, slow analyses, may damage the biofilm
Confocal laser scanning microscopy (CLSM)	Able to produce 3D images of biofilm efficiently monitoring bacterial growth, metabolic activity and gene expression in biofilm, and allows studying the physio-chemical and biochemical aspects of biofilm microenvironments	Overlapping of the fluorescence signals of the auto-fluorescence biomolecules and fluorophores, limitations over the number of the fluorescence filters combinations and unsuitable for use with opaque and very thick biofilm
Atomic force microscopy	Has a high resolution and it can be used in vivo studies	Sample dehydration during the examination which may affect the accuracy of the extracted biofilm information
X-ray microscopy	High resolution, simplicity in preparing the samples and maintenance of hydration of biofilm sample	Unsuitable for thick biofilms (<10 μm), and a destructive mode of analysis
Raman microscopy	Can examine the spatial distribution of microorganisms in the biofilm matrix in a non-invasive way. Capable of yielding spatially resolved chemical information of the biofilm	Restricted to infrared wavelength. There is also a lack of spectral database of microbes without which we cannot differentiate between species of microbes
Hoffman modulation contrast microscopy (HMCM)	Non-invasive microscopic technique, ability of HMCM to produce 3D image, HMCM has other advantages such as high contrast resolution, suitability to use with dense biofilm and no requirements for sample preparation	No notable drawbacks
Differential interference contrast microscopy (DICM)	Rapid way for monitoring biofilm and it has the capacity to produce 3D images of in situ biofilm	It is fragile and sensitive to heat. Uses expensive quartz Wollaston prisms. The signal is reduced by the presence of the polarizer. Image contrast is reduced by the presence of birefringent materials. Varying ellipticity of polarization of laser light causes fluctuations in brightness of produced DIC images
Environmental scanning electron microscopy (ESEM)	Can analyze hydrated biofilms	Cannot be used for in vivo and on-line monitoring systems. Poor distinguishing between small cells and the texture of the substrate in a biofilm with random topography
Digital time-lapse microscopy	Can study the effect of membrane surface properties on initial adhesion of bacteria, effect of nutrients and flow conditions on deposition of microorganisms on RO membrane	Observed area in the flow cell is very limited which may not give an accurate representation for the case. Limitation of depth in the flow cells restricts the flow in the cell to laminar conditions
Spectroscopic techniques		
Fourier transform-infrared (FT-IR) spectroscopy	Required volume of sample is very small (range of ng-μg), can analyze samples of different phases and identify if microorganisms are dead or alive	Can only detect thin biofilms of the order of 1 μm and for accurate analysis, a complete library of the spectra for each microorganism is required
Bioluminescence	Can identify characteristics of biofilm such as bacterial biomass, cellular activity and gene expression in genetically modified bacteria	Confined to environments possessing microorganisms that are naturally or genetically modified to emit light under the effect of biochemical reactions
Nuclear magnetic resonance (NMR) spectroscopy	Non-destructive and non-invasive. Can monitor growth state of microorganisms in biofilm, the architecture of the biofilm and the detachment rate of the biofilm under starvation conditions as well as effect of biofilm on the hydrodynamics of the surrounding liquid	Low signal/noise ratio, long time required for data acquisition and the quality of the produced images by NMR is affected by the surface curvature of the biofilm. Expensive technique because isotopes required in NMR spectroscopy are naturally scarce
Pressure drop measurements	Cost effective technique for monitoring early stage biofouling in membrane systems	Cannot specifically detect biofilm formation on the membrane as pressure drop can be due to factors other than biofouling too

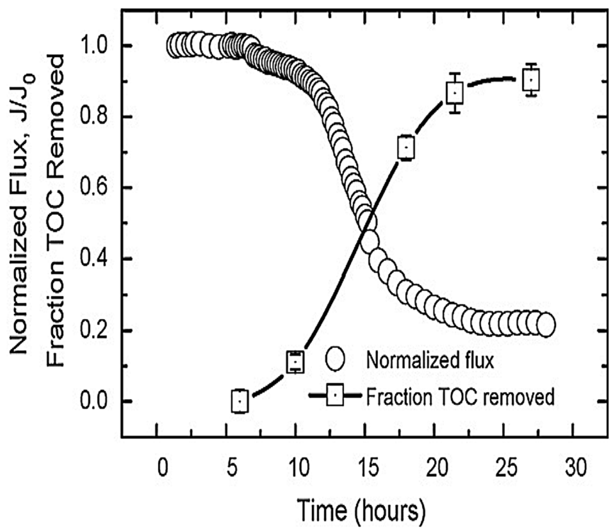


Fig. 7 Permeate flux and TOC removal upon growth of biofilm on an RO membrane (Herzberg and Elimelech 2007)

7. Serious decline in the quality of permeate: This is because of all the factors previously listed.
8. Higher treatment costs: This results from high energy requirements, cleaning demand, and membrane replacement.

Permeate flux decline exhibits two phases; initial rapid decline followed by a more gradual decay. The rapid decline takes place in the early attachment stage while the slow decline occurs during the plateau phase. In the presence of bacteria, the higher the permeate volume required, the greater the flux decline is observed, Fig. 7. System pressure will increase to compensate for the flux decline and this will add more treatment costs. The main reason for the decline in flux or salt rejection is that bacterial cells hinder the back diffusion of salts by secreting EPS which then increases hydrolytic resistance of the membrane. In particular, EPS fouling only showed salt rejection decrease by 2%, but with dead cells, reduction could reach up to 5–6%. Membrane biodegradation is another reason for the decrease in salt rejection in RO cellulose acetate modules (Matin et al. 2011; Herzberg et al. 2009).

Gradual accumulation of dissolved substances retained by the membrane at the raw waterside initiates concentration polarization phenomenon. The increase in hydraulic resistance also results in reducing permeate flux and enhancing concentration polarization which causes a decrease in salt rejection (Matin et al. 2011). Concentration polarization occurs when the salt concentration near the membrane surface exceeds the salt concentration in the bulk solution because of flow of water through the membrane and rejection of salts (Flemming 1997). We have four key factors to determine the magnitude of concentration polarization: the boundary-layer thickness, the

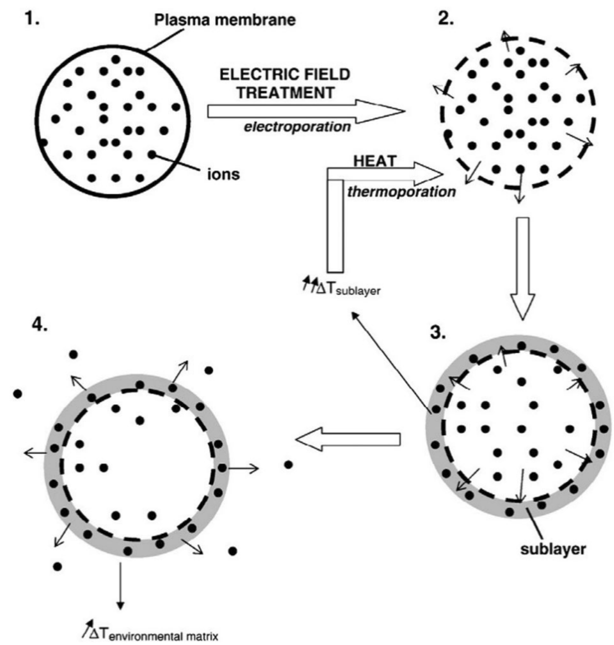


Fig. 8 Death of a cell caused by PEF (Guyot et al. 2007)

permeate flux, the membrane development and the solute diffusion coefficient in the boundary-layer fluid. Concentration polarization results in the following effects: reduces the net driving pressure differential across the membrane, thus, lowering the permeate flow rate, increases salt flow across the membrane, and increases precipitation that causes membrane scaling (Qureshi et al. 2013).

Concentration polarization strongly affects the performance of the separation process. First, concentration changes in the solution reduce the driving force within the membrane, hence, affecting the useful flux/rate of separation. In the case of pressure driven processes, this phenomenon causes an increase in the osmotic pressure gradient of the membrane reducing the net driving pressure gradient. In the case of electromembrane processes, the potential drop in the diffusion boundary layers reduces the gradient of electric potential in the membrane. Lower rate of separation under the same external driving force means increased power consumption (Baker 2012).

A case study showed that, because of an additional hydraulic resistance of the biofouling layer, Water Factory 21, Orange County, CA, operates at about 150% of their initial operating pressure (roughly 200 psi). It was observed that the $1 million membrane inventory lasted only for 4 years instead of its theoretical life-span of 8 years. This amounts to an added cost of $125,000 per year due to biofouling (Flemming 1997; Flemming et al. 1994). Maddah et al. showed in their membrane cost study analyses that integrated UF-RO membranes have the lowest treatment cost of $0.3/m^3 compared to MF-RO and MBR types

(~\$0.5/m^3) since UF membranes can control foulants before they reach at the RO module and damage it. Therefore, fouling costs were eliminated in UF-RO reducing the overall treatment cost for the UF-RO modules (Maddah and Chogle 2015).

Control and remediation

After detection and monitoring of biological matter that is responsible for forming biofilms, the next stage is successful enactment of remediation techniques for controlling biofouling in RO systems. Techniques employed for controlling biofouling include the following:

Membrane cleaning

Membrane cleaning involves physical cleaning, backwashing, chemical cleaning, removal of organic films, slimes, and biological fouling. It contributes to 5–20% of the operating cost. Chemical cleaning agents are commercially available and they are included in six categories: alkalis, acids, metal chelating agents, surfactants, oxidation agents, and enzymes. The most effective combination is enzyme–anti-precipitant–dispersant and bactericidal agent with an anionic detergent for cellulose acetate RO membranes. Another noteworthy combination is chelating agent surfactant with alkali for polyamide RO membranes (Matin et al. 2011).

Cleaning chemicals should be used wisely in RO membranes as they could be harmful to the membrane material since frequent cleaning may cause conditioning or hardening of foulant layers (Baker and Dudley 1998). Moreover, cleaning techniques are employed after biofouling has already occurred. Therefore, since prevention is better than cure, focusing on feed pretreatment is the optimal approach to prevent biofouling repercussions.

Feed pretreatment includes acid dosing for pH control, coagulation and flocculation, media filtration, chlorination, ozonation, UV radiation, addition of antiscaling compounds or inhibitors, cartridge filters, activated carbon adsorption, etc. Practically, in RO systems disinfection is done by chlorine and copper sulphate while coagulation is carried out by alum (El Aleem et al. 1998).

Disinfection

Biofouling cannot be eradicated by pretreatment alone. Even if 99.99% of all bacteria are eliminated by pretreatment, a few surviving cells will enter the system and multiply. Biofouling occurs even after significant disinfection with chlorine. In the Middle East, about 70% of the seawater RO plants suffer from biofouling problems which can be resolved by the application of several physical and chemical disinfection techniques which are categorized and summarized in Table 6 (Matin et al. 2011; Al-Juboori and Yusaf 2012; Young 1999).

Biocides are materials and substances that are used for the purpose of feed pretreatment and are categorized as oxidizing and non-oxidizing biocides. Oxidizing agents include chlorine, bromine, chloramine (NH_2Cl), chlorine dioxide (ClO_2), hydrogen peroxide, peroxyacetic acid, hypochlorous acid (HOCl), and ozone while non-oxidizing agents include formaldehyde, glutaraldehyde, quaternary ammonium compounds, etc. Oxidizing agents are applied to industrial water treatment plants, but are incompatible with polyamide RO membranes since they may break down humic acids into smaller components that serve as nutrients to bacteria. On the other hand, non-oxidizing agents are more relevant to industrial wastewater treatment plants since they are more compatible with RO membranes. It is recommended to avoid using low levels of biocides on microbes because continuous low dose rates often cause microbial resistance (Matin et al. 2011).

Chlorine is another biocide which is used for chlorination; another technique that is not viable anymore because it is found that chlorine is responsible for the degradation of humic acids to smaller molecules that are used as nutrients to bacteria. Another reason is related to the aftergrowth mechanism in which there is a sharp increase in bacteria after dechlorination with sodium metabisulfite (SBS) since surviving bacteria utilize the degraded molecules and use them as nutrients (Abd 1998). However, disinfectants like chloramine and copper sulfate would be excellent substitutes for chlorine. Stopping chlorination/dechlorination altogether is the most recommended approach to achieve more successful operations and improved performances. Intermittent or shock dosing chlorination is an excellent alternative to plants which operate without chlorine; it is suggested to chlorinate for 6–8 h per week with a residual chlorine level of 1 mg/l (Saeed et al. 2000). Similarly, shock dosing is also performed by using sodium bisulphite ($NaHSO_3$) for an exposure time of 30 min at a concentration of 500 ppm with kill rates up to 99% for seawater microflora (Baker and Dudley 1998).

On the contrary, under physical methods we have electrical techniques used for water disinfection that include electro-chemical techniques and pulsed electric field (PEF). Electro-chemical techniques can be categorized into two groups, namely, methods that use direct electrolysers which interact directly with microbes, and other methods that use mixed oxidant generators producing oxidizing species for damaging microbes. PEF as seen in Fig. 8 is a disinfection technique that involves maintaining the suspension of microorganisms between electrodes and

Table 6 Summary of disinfection techniques (Matin et al. 2011; Al-Juboori and Yusaf 2012; Young 1999; Kim et al. 2009)

	Disinfection	Advantages	Disadvantages
Chemical	Chlorine (HOCl, NH_2Cl, ClO_2) (Matin et al. 2011)	Initial removal of biofouling prior to dechlorination, relatively low cost, less or no damage to membrane	Dechlorination may enhance biofouling, chlorination gives carcinogens (THMs, HAAs), chemically corrosive, chlorite toxicity, low efficiency
	Ozone (Matin et al. 2011)	High oxidation, ideal when combined with GAC	Costly and generates carcinogens (bromate), very small half-life
Physical	UV (Matin et al. 2011; Al-Juboori and Yusaf 2012)	No by-products, enhanced performance when combined with sodium hypochlorite, easy installation and maintenance	Scale formation and may produce mutagenic components
	Sand filtration	Low installation and operation cost	Low bacteria removal efficiency
	Electrical (Al-Juboori and Yusaf 2012)	Lower energy requirement, do not produce a new generation of microbes that are tolerant to the treatment	It may produce mutagenic components in the treated water, cathode fouling
	Ultrasound (Young 1999)	Can be combined with other techniques to enhance performance, used for solutions having suspended solids	High cost, requirement of cooling processes

subjecting them to a high intensity of electric field for a short period to degrade the microorganisms directly by decomposing DNA or RNA of their cells (Al-Juboori and Yusaf 2012).

The advantages of electrical disinfection methods include lower energy requirement, which unlike thermal techniques, do not require energy expenditure in the form of cooling. In addition, these methods do not produce a new generation of microorganisms that are tolerant to electrical treatment. However, it may produce mutagenic components in the treated water as well as cathode fouling (Al-Juboori and Yusaf 2012).

Ultrasound Techniques are employed primarily as replacements for UV light and chlorination treatments for water disinfection, but can also be used for performance enhancement. These techniques include acoustic cavitation phenomenon (Fig. 9) that occurs due to the fall of the ambient pressure under the saturated vapor pressure of the liquid because of ultrasound waves passing through the liquid. There is an oscillation of pressure due to ultrasound waves; the positive swing of pressure is called compression period and the negative swing is called rarefaction period. Formation of voids takes place during the rarefaction period, while the collapse of bubbles takes place during the compression period (Young 1999).

Cavitation can be homogeneous, where the generation of bubbles is due to interaction between liquid and vapor, or heterogeneous, where the interaction is between solid, liquid, and vapor phases simultaneously. The surface tension of the liquid at the nucleation sites (where cavitation occurs) is weak which allows the negative pressure of sound waves to rupture the liquid and generate bubbles. The bubbles forming in the liquid as a result of irradiation may collapse either gently (stable cavitation) or violently (transient cavitation). Transient cavitation exists normally

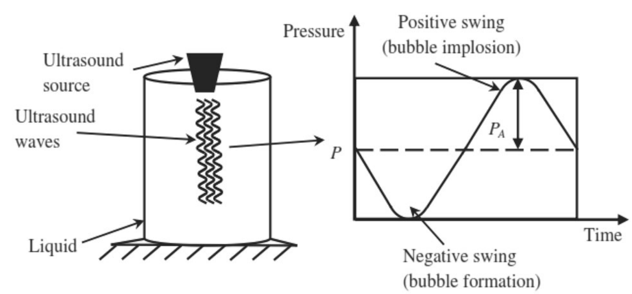


Fig. 9 Acoustic cavitation process (Young 1999)

for 1 cycle of the sound pressure in which bubbles expand to at least double their size and collapse severely often disintegrating into small bubbles. Whereas, stable bubbles can oscillate for more than one cycle of sound pressure and grow due to mass diffusion through the bubble (Young 1999).

Mechanism of ultrasound involves three stages. First, the mechanical effects stage results from the cavitation phenomena. Second, the chemical effects of cavitation phenomena occur, which involve the generation of free radicals in the medium. Third, heat effects represented by the generation of localized hot spots developed by rapid explosion of the bubbles (Young 1999).

It was observed that mechanical effects play the main role in destroying microorganisms while chemical effects and heat effects play only a supporting role. Implosion of bubbles generates mechanical effects such as high pressure, turbulence due to liquid circulation, and shear stresses. Micro-streaming resulting from bubble oscillation can generate stresses that have the potential to rupture microorganisms. It was proposed that in ultrasound treatment, cell rupture occurs due to exposure of cells to viscous dissipative eddies that generate from the shock waves of bubble collapse. The main cause of cell disruption in ultrasound treatment was later confirmed to be the collapse pressure that results from bubble implosion (Young 1999).

pH adjustment

pH adjustment is recommended to control adhesion of microbes on the RO membrane. pH can either be increased by addition of a strong base like NaOH or decreased by addition of a strong acid like HCl. The addition of an acid is not recommended as it can lead to corrosion of the membrane. It is also known that organic fouling is usually accelerated with decrease in pH and increase in divalent cation concentration. In low pH and high divalent cation concentration, charge property of organic matters diminishes through the neutralization of functional groups as well as organic-calcium complexation. Moreover, it has been found that increasing pH of feed water is not as helpful as initially presumed. Feed water pH affects both the charge properties of bulk organic foulants as well as the interfacial interaction between organic foulants and membrane surfaces. The former leads to the formation of thick and dense fouling layers on the membrane surface due to the favorable multi-layer accumulation of organic foulants. The latter results in the reduction of electrostatic repulsion between organic foulants and membrane surfaces leading to accelerated accumulation of the foulants on the membrane surface (Al-Juboori and Yusaf 2012).

The effect of pH is noticeable only when the feed water has low ionic concentration. Increasing pH in such a feed can lower the flux decline rate. However, when the ionic concentration of feed water is high, there is a negligible change in flux decline rate. As reverse osmosis is used for desalination of seawater, variations in pH are not beneficial since seawater has high ionic concentration. Thus, feed water pH is not a significant factor affecting organic or biological fouling during seawater desalination (Herzberg and Elimelech 2007; Baek et al. 2011).

Membrane surface modification

Surface modification techniques are employed to improve certain membrane characteristics such as surface roughness, surface charge and membrane hydrophilicity.

Surface roughness as discussed earlier, increases microbial adhesion due to higher surface area as compared to a smooth surface. Moreover, the peaks and troughs of rough surfaces provide higher frequency of susceptible sites for microbial adhesion. This problem can be considerably reduced by smoothening the membrane surface with the application of a thin layer of polymer. Thin polymeric film is physically coated on the membrane surface. This polymer can not only possess characteristics such as high hydrophilicity, but also can be reactive in nature (Malaisamy et al. 2010). The surface roughness of RO membranes is also positively correlated with colloidal fouling (Kang and Cao 2012).

Most species of bacteria are negatively charged and hence, to reduce microbial adhesion, the theory of making membranes negatively charged was proposed. The electrostatic repulsion existing between microorganisms and the negatively charged membrane will inhibit adhesion and hence, biofouling (Kang and Cao 2012).

Increasing the hydrophilicity of a membrane leads to decrease in the attachment of microorganisms to the membrane surface as the hydrophilic membrane favors interaction with water molecules in lieu of microorganisms. In other words, hydrophobic membranes prefer interacting with microorganisms resulting in greater microbial adhesion. The hydrophilicity of a membrane can be increased by physically coating the membrane surface with a thin polymeric film.

Improvement of membrane surface is possible by adding active organic modifiers into trimesoyl chloride (TMC) or m-phenylenediamine (MPD) solution. Currently, TMC and MPD are the most commonly used active monomers to form functional polyamide layer in RO membrane. An earlier study showed that a novel prepared composite RO membrane from 5-isocyanato-isophthaloyl chloride (ICIC) and MPD had favorable hydrophilicity and smoother surface, and therefore ICIC-MPD membrane showed better resistance to fouling (Kang and Cao 2012).

Interestingly, Yang et al. (2011) synthesized a modified RO membrane which was chemically grafted with poly-(sulfobetaine) zwitterionic groups for surface development. The modified RO membranes exhibited superior antifouling performance against E. coli and showed long-term operation compatibility because the modifiers were covalently connected with the membrane surface. Practically, the coating layer must be synthesized sufficiently thin to maintain the water flux and water permeability as high as possible (Kang and Cao 2012).

Malaisamy et al. (2010) used polymeric films for membrane modification to produce acrylic acid (AA) modified and [2-(acryloyloxy)ethyl] trimethyl ammonium chloride (AETMA) modified membranes. AETMA-modified membranes, in addition to having higher flux than AA-modified membranes, possess antibacterial properties that minimizes the biofoulant growth (Hyun et al. 2006; Lee et al. 2010; Yang et al. 2010) Moreover, AA-modified membranes, when fouled even with trace levels of bacteria, cannot prevent their growth. Hence, AETMA-modified membranes are most desirable for increasing hydrophilicity along with anti-bacterial behavior (Malaisamy et al. 2010). Thin-film polyamide composite RO membranes can be modified by the addition of aliphatic and aromatic groups.

Khan et al. (2010) have found that the addition of aliphatic hydrocarbon groups on the polyamide layer of RO membranes increased biofouling compared to the addition of aromatics.

Moreover, RO membrane from 3-monomethylol-5,5-dimethylhydantoin (MDMH) is characterized with improved surface hydrophilicity as well as substantial biofouling prevention which is confirmed by testing the membrane with Escherichia coli (E. coli) as a model for microorganism foulants. Not only this, MDMH-modified RO membrane offered substantial chlorine resistance making this membrane ideal in chlorine resistant and anti-biofouling applications (Kang and Cao 2012; Wei et al. 2010a, b).

Hybrid organic/inorganic RO membrane process is carried out by coating RO membranes with inorganic particles by direct deposition or via interfacial polymerization process. Inorganic particles include photocatalytic titanium dioxide (TiO_2), SiO_2, Zeolite A, and silver nanoparticles (Kang and Cao 2012). Nanomaterials also include chitosan, aqueous fullerene nanoparticles and carbon nanotubes (Matin et al. 2011).

Hybrid membrane with TiO_2 nanoparticles can be introduced as a commercial RO membrane and they are capable of increasing the water permeability by 20%. Fig. 10 confirms that the combination of TiO_2 and UV light is the optimal choice for decimation of E. coli population. Silver compounds are strong bacterial growth inhibitors since silver ions can react with thiol (−SH) groups in microbial cells for the inactivation of bacterial growth. Bacterial colonies were found to be at least 98% less in coated silver nanoparticles substrates compared to the surrounding uncoated regions (Matin et al. 2011).

Furthermore, hybrid membranes are very promising in commercial use since they are capable of enhancing permeability characteristics and antifouling as discussed earlier. Besides, they are characterized with self-cleaning properties. For example, depositing TiO_2 nanoparticles onto aromatic polyamide RO membrane surfaces showed an excellent antibacterial fouling potential and this is confirmed by Madaeni and Ghaemi (Madaeni and Ghaemi 2007) who created a self-cleaning RO membrane using TiO_2 as a coating. Moreover, hybrid zeolite-polyamide membranes (Jeong et al. 2007) showed enhanced surface hydrophilicity with greater negative charge and lower roughness which implies that zeolite-polyamide membranes have a strong potential to be used as antifouling membranes (Kang and Cao 2012).

Rana et al. (2011) added 0.25 wt% of silver salt into aqueous MPD phase to improve membrane surface hydrophilicity and achieve better anti-biofouling property. However, deposited inorganic particles onto RO membrane surface may face a problem of loss or leaching in long-term operations. It is worth mentioning that modifiers with

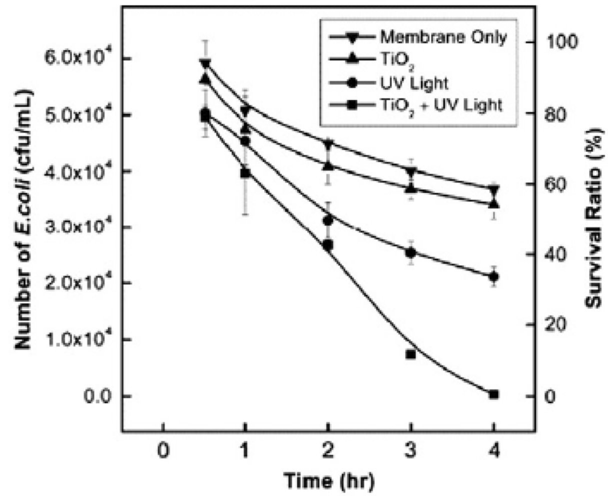


Fig. 10 Ratio of flux to their initial values during fouling experiment (Matin et al. 2011)

chemically covalent bonds with membrane can withstand longer than modifiers with physical bonds such as van der Waals attractions, hydrogen bonding or electrostatic interaction (Kang and Cao 2012; Rana et al. 2011).

Biochemical action

Biochemical materials like enzymes and bacteriophages can be used to alleviate detrimental effects of biofouling. While EPS may consist of exopolysaccharides, proteins, glycol-proteins, released nucleic acid, phospholipids and other surfactants, polysaccharides and proteins are the two main components of EPS. Hence, enzymatic action is directed towards them. These enzymes are of two main types, namely, polysaccharide lyases and hydrolases. For proteins, there are degrading enzymes called proteases, which are categorized as exopeptidases and endopeptidases. Along with polysaccharide lyases, hydrolases, and proteases, bacteriophages are also employed for biochemical control of biofouling (Al-Juboori and Yusaf 2012; Richards and Cloete 2010).

Conclusion

Microorganisms such as bacteria, fungi and yeast are major enemies of desalination plants that involve reverse osmosis modules. Biofilm formation occurs in a series of events that are conditioning (adsorption), transport and attachment of microbes, growth and detachment. In the growth stage, extracellular polymeric substances are produced continuously to provide nutrients to bacteria and offer defense against biocides. It has been observed that increasing pH of feed water would reduce the permeate flux decline rate.

Hoffman modulation contrast microscopy and Fourier transform-infrared spectroscopy are determined to be the best microscopic and spectroscopic techniques, respectively, for the detection of biofouling in reverse osmosis membranes as their disadvantages are either negligible or can be minimized. Biofouling causes permeate flux and quality decline, membrane biodegradation, and an increase in salt passage through concentration polarization. Biofouling also increases desalination treatment costs by up to 50% of the total costs due to membrane life shortening, and higher energy requirement.

Biofouling can be effectively reduced by two different pretreatment techniques that are disinfection and pH adjustment. Chlorination and ozonation are some chemical disinfectants while UV, sand filtration, electrical treatment, and ultrasound technique are physical disinfection agents. The problem with chlorination is that surviving bacteria will utilize sodium metabisulfite for nutrition after dechlorination and therefore it is not an ideal choice to prevent biofouling. Intermittent or shock dosing chlorination is an excellent alternative to plants which operate without chlorine. Shock dosing is also performed by using sodium bisulphite ($NaHSO_3$) with kill rates up to 99% for seawater microflora. Membrane surface modification is the best technique for the prevention of biofouling as it increases membrane hydrophilicity, decreases surface roughness, and may restrict microbial adhesion by electrostatic repulsion. Hybrid organic/inorganic RO membranes are promising in dealing with biofouling since deposited inorganics such as photocatalytic titanium dioxide (TiO_2), SiO_2, Zeolite A, and silver nanoparticles are excellent in reducing microorganism populations. The last option to handle biofouling once it has already occurred is membrane cleaning which contributes to 5–20% of the operating cost. Membrane cleaning involves physical cleaning, backwashing, chemical cleaning, removal of organic films, slimes, and biological fouling.

Biofouling poses a serious threat to efficient desalination processes. However, this paper gives a glimpse of the different techniques that would overcome these challenges. The authors believe that each remediation technique may have pros and cons, and hence further research is needed to identify the perfect approach for complete eradication of biofouling.

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